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N786

B5A AC A1R214H A1R314C1A A1R314C1C  
A1R314C1D A1R314C1E A1R314C1F A1R314C6  
A1R400 A1R404 A1R420 A1R429X A1R462 A20T14  
A20T15 A20T17  
B8F FBF FBG F1D2  
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(54) Heat-activated labels

(57) A heat-activated label is provided having a substrate and an adhesive layer provided on the back side of the substrate, in which an adhesive used in the adhesive layer contains an ethylene- $\alpha$ -olefin copolymer obtained by copolymerization of ethylene and an  $\alpha$ -olefin in the presence of a metallocene compound-containing catalyst. The label (1) may be stuck to a previously moulded article (11) or a moulded article may be produced in which the label is in the mould 23.

Fig. 1

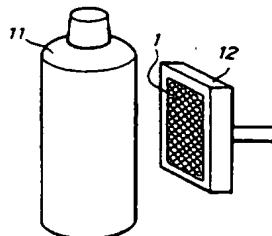


Fig. 2a

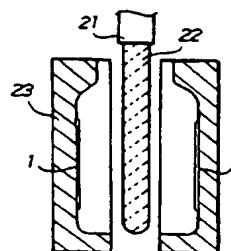
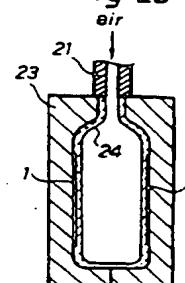


Fig. 2b



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(58) Field of Search

UK CL (Edition O ) BSN

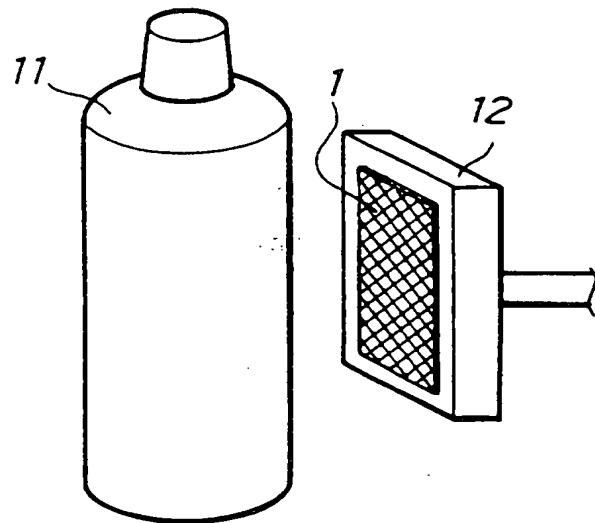
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123/02 123/04 123/08 123/16 , G09F 3/04

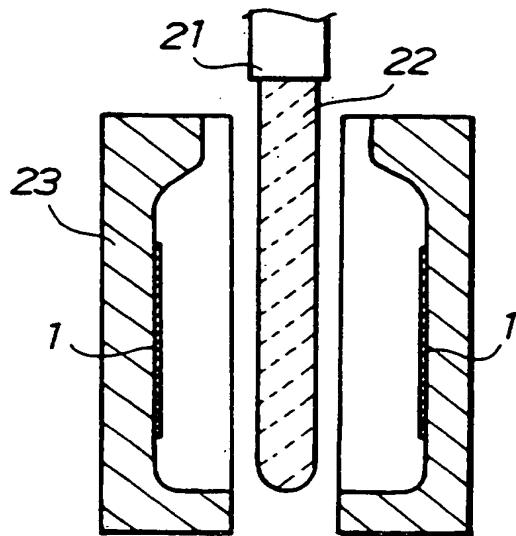
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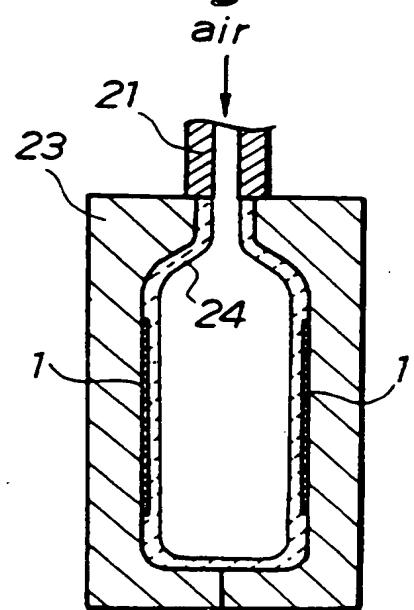
*Fig. 1*



*Fig. 2a*



*Fig. 2b*



**2310398**

Title Of The Invention

HEAT-ACTIVATED LABEL AND  
PROCESS FOR PRODUCING LABELED MOLDED ARTICLE

BACKGROUND OF THE INVENTION

Field of the Invention:

The present invention relates to a heat-activated label exhibiting high adhesion and a highly productive process for producing a labeled molded article.

Description of Related Art

Heat-activated labels having an adhesive layer mainly comprising an ethylene copolymer are known. In order to lower the temperature at which the adhesive action of these labels is activated, it has been a common practice to lower the melting point by increasing the proportion of comonomers in the ethylene copolymer. Components copolymerizable with ethylene which are generally used include vinyl acetate, ethyl acrylate, acrylic acid, and methacrylic acid (see, for example, Japanese Patent Laid-Open No. 2-258556). Sufficient reduction in melting point has been achieved in this manner to obtain satisfactory low-temperature adhesion.

However, when the label is to be adhered to an olefin resin molded article, compatibility to the molded article is reduced as the melting point decreases with an increase of the comonomer only to give poor adhesion.

In order to improve compatibility to an olefin resin, it has been proposed to use, as an adhesive component, an ethylene- $\alpha$ -olefin copolymer which is obtained by copolymerizing ethylene and an  $\alpha$ -olefin comonomer using a conventional Ziegler-Natta catalyst. However, when the ethylene- $\alpha$ -olefin copolymer is prepared using an increased amount of an  $\alpha$ -olefin in order to reduce its melting point to 50 to 80°C, the resulting copolymer contains an increased proportion of low-molecular components. Such a copolymer has stickiness or causes blocking, making handling of the resulting heat-activated adhesive label difficult.

Labeled molded articles have been conventionally obtained by in-mold labeling<sup>2,3</sup> in which molding is performed in a mold into which a label has previously been placed. The problem associated with in-mold labeling is that the mold must be cooled for a sufficiently long time because the thickness of the label added to the thickness of the molded article reduces the cooling efficiency. Also, if the temperature at

which the molded article is removed from the mold is too high, the label develops blisters. Therefore, the molding cycle for in-mold labeling is generally long, causing a productivity problem in the production of labeled molded articles.

In order to improve productivity in in-mold labeling, one approach generally taken is to set the molding temperature (resin temperature) low to decrease the quantity of heat possessed by the molded article in the mold and to shorten the mold cooling time, i.e., to remove the molded article while at a relatively high temperature in the molding cycle.

When a heat-activated label using the above-described conventional adhesive having a reduced melting point is used to assure sufficient adhesion at a low molding temperature, the adhesive is re-melted by so-called heat-backing. This, however, causes blisters because of the short cooling time and the high temperature at the time of removal from the mold. On the other hand, if the adhesive used has a high melting point so as to prevent blistering even at a high removal temperature, it exerts reduced adhesion at a low molding temperature.

Various proposals have been made for in-mold labeling or in-molding labeling techniques in order to

suppress blistering even when molding is performed in a short cycle.

For example, Japanese Patent Laid-Open No. 5-8289 discloses a process for producing a labeled hollow container wherein the adhesive layer of the label comprises a mixture of at least two resins having different melting points satisfying specific conditions.

According to the disclosure, the resin having a higher melting point undergoes setting at a high temperature and therefore acts to inhibit blistering even when the container is removed from the mold at a high temperature, while the resin having a lower melting point melts at a low temperature and therefore serves to maintain adhesiveness at a low molding temperature.

Even with such an adhesive, the high-melting component still interferes with adhesion of the label to the molded article in low-temperature molding, and the low-melting component still causes blisters in high-temperature removal from the mold. Thus, the adhesive causes insufficient improvements in productivity.

Japanese Patent Laid-Open No. 5-24048 discloses a process for producing a labeled container wherein a labeled container taken out of the mold after a short cooling time is quenched to a temperature below the

crystallizing or softening point of the adhesive immediately after removal from the mold.

It is suggested that the quenching can be carried out by spraying water or cooling air or by cooling in a mold provided for supplementary cooling. According to this proposal, although development of blisters is suppressed, a cooling system should be incorporated into a general molding machine, making the production equipment larger and more complicated, which ultimately results in a reduction in productivity and an increase in cost.

Additional proposals have been made aiming at improvement in adhesion or prevention of blisters (e.g., Japanese Patent Laid-Open Nos. 4-4121, 4-4130, 4-29184 and 4-359284 and Japanese Patent Publication No. 6-70736), but none of them succeeded in achieving sufficient improvements in adhesion or sufficient reduction in the molding cycle.

Further, a labeler adhesion method has also been used for producing a labeled molded article, in which a heat-activated label is activated by heating with a labeler equipped with a heating system and the heated label is stuck to the surface of a previously prepared molded article while the adhesive layer of the label retains adhesiveness (remains activated).

In the labeler adhesion method, the adhesive to be used in the heat-activated label should have a low melting point in order to be activated at a temperature that is as low as possible. If the melting point is reduced by using the above-described comonomers, such as vinyl acetate, ethyl acrylate or (meth)acrylic acid, the adhesive will have a broadened range of melting point and increased polarity. The label then tends to cause blocking before being activated. In other words, the labels placed one on another stick to each other and are difficult to feed individually.

The labeler adhesion method also has the following disadvantages: the label has weak adhesion after it is adhered to the molded article and needs time for the adhesive to set; the label edges tend to release from the adherent when the labeling speed is high, and cooling is insufficient.

In brief, it has been impossible for conventional heat-activated labels to obtain high adhesion to various molded articles, including olefin resin molded articles, at a low temperature and to produce a labeled molded article at a high production speed by means of a conventional molding machine. When adhered by means of a labeler equipped with a heating system, the conventional heat-activated labels involve the problem of

blocking and poor adhesion after labeling.

#### SUMMARY OF THE INVENTION

Accordingly, one object of the present invention is to provide a heat-activated label which exhibits excellent adhesion at a low temperature and does not cause blocking.

Another object of the present invention is to provide a process for producing a labeled molded article which can be carried out with a greatly reduced molding cycle, even with conventional equipment, for in-mold labeling.

A further object of the invention is to provide a process for producing a labeled molded article which is free from such problems as label release from a molded article when adhered by means of a labeler.

These and other objects of the present invention have been satisfied by the discovery of a heat-activated label which comprises a substrate and an adhesive layer provided on the back side of the substrate, wherein the adhesive used in the adhesive layer comprises an ethylene- $\alpha$ -olefin copolymer obtained by copolymerization of ethylene and an  $\alpha$ -olefin in the presence of a catalyst comprising a metallocene compound.

The present invention also provides the above-described heat-activated label, in which the melting point of the ethylene-a-olefin copolymer is from 50 to 80°C, and the label is to be heated and then stuck to a previously prepared molded article.

The present invention also provides the above-described heat-activated label, in which the melting point of the ethylene-a-olefin copolymer is from 70 to 100°C, and the label is to be inserted into a mold for in-mold labeling.

The present invention further provides a process for producing a labeled molded article, which comprises heating a heat-activated label in accordance with the present invention and sticking the heated label onto the surface of a previously prepared molded article (hereinafter referred to as a first process).

The present invention further provides a process for producing a labeled molded article, which comprises producing a molded article by using a mold in which a heat-activated label in accordance with the present invention has previously been inserted (hereinafter referred to as a second process).

The present invention also provides the above-described processes for producing a labeled molded article, in which the labeled molded article is a blown

container, wherein at least the side of the container having the label thereon comprises a polyolefin resin.

The heat-activated label according to the present invention has excellent adhesion at low temperatures and does not cause blocking.

The processes of the present invention for producing a labeled molded article make it feasible to markedly reduce the molding cycle even when using a conventional in-mold labeling machine and does not cause such problems as label release when labeling is conducted by means of a labeler.

#### BRIEF DESCRIPTION OF THE FIGURES

A more complete appreciation of the present invention and many of the attendant advantages thereof will be readily obtained as the same becomes better understood by reference to the following detailed description when considered in connection with the accompanying drawings, wherein:

Fig. 1 schematically shows an embodiment of the first process for producing a labeled molded article according to the invention.

Figs. 2(a) and 2(b) schematically illustrate an embodiment of the second process for producing a labeled molded article according to the invention, in

which Fig. 2(a) shows the state before blowing, and Fig. 2(b) shows the state after blowing.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The heat-activated label of the present invention comprises a substrate and an adhesive layer provided on the back side of the substrate wherein the adhesive used in the adhesive layer comprises a specific resin.

Within the context of the present invention, the term "heat-activated label" is intended to mean a label (preferably with various pieces of information printed on the back side thereof), having an adhesive layer that is melted and shows tack( i.e., is activated), on heating at a temperature above room temperature but is in a solidified state with no tack before heat activation(i.e., at room temperature), and therefore can be stored and handled without using release paper or other release agents.

The heat-activated label of the present invention can be adhered to a molded article, for example, by a labeler adhesion method in which a heat-activated label is adhered to a previously prepared molded article and an in-mold labeling method in which a resin is molded in a mold in which a heat-activated label has

been inserted, and the heat-activated label is activated by the heat of the resin and adhered to the molded article.

Suitable substrates for use in the present invention include papers, synthetic papers (e.g., stretched films comprising a resin and an inorganic filler), stretched or unstretched plastic films (e.g., stretched or unstretched films of polyethylene resin, polypropylene resin, polyester resin, polystyrene resin or polyamide resin), and nonwoven fabrics.

When the heat-activated label is used in in-mold decoration, especially for the production of thin-wall articles, the substrate is preferably made of a material having such dynamic physical properties that it shrinks at a percent shrinkage near that of the molded article or it can follow the shrinkage of the molded article (e.g., a relatively small modulus). More specifically, an unstretched film of a polypropylene resin or a polyethylene resin is preferably used.

The substrate may have a structure with several divided layers or may be a composite laminate of different materials.

Where the substrate is disposed as an outermost layer, it is preferably subjected to antistatic treatment. An antistatic treatment can be carried out by

applying a known antistatic agent to the surface or adding an antistatic agent to the material forming the substrate. The amount of the antistatic agent, when added to the substrate forming material, is arbitrary but is preferably chosen to give a concentration that will not impair the adhesion to the adhesive layer and the printed layer described hereinbelow.

The surface of the substrate to be printed is preferably subjected to a surface treatment for improving printability, such as a corona discharge treatment.

A heat insulating material, such as a foamed plastic, may be used as a substrate or part of a substrate in order to prevent the heat, once applied to the adhesive layer for activation, from being conducted to other members in contact with the label, such as a mold in in-mold labeling or a drum of a labeler in post labeling. Such loss of heat results in reduced activation efficiency.

The substrate preferably has a thickness of 40 to 200  $\mu\text{m}$ , more preferably 60 to 150  $\mu\text{m}$ . If the thickness is smaller than 40  $\mu\text{m}$ , the label has poor bending stiffness and is difficult to stick to a molded article or to insert into the mold cavity. A substrate thickness exceeding 200  $\mu\text{m}$  tends to cause blisters, especially in in-mold labeling.

Printing of various pieces of information on the substrate can be carried out using any conventional printing method, such as gravure printing, offset printing, flexographic printing, and screen printing, without any particular restriction.

The specific resin which can be used as an adhesive of the heat-activated label of the present invention is an ethylene- $\alpha$ -olefin copolymer obtained by copolymerizing ethylene and an  $\alpha$ -olefin in the presence of a catalyst comprising a metallocene compound (hereinafter referred to as MPE).

Suitable  $\alpha$ -olefins which can be used in MPE include those having 3 to 20 carbon atoms, such as propylene, 1-butene, 1-pentene, 1-hexane, 1-octene, 1-heptene, 4-methyl-1-pentene, 4-methyl-1-hexene, 4,4-dimethyl-1-pentene, octadecene and isooctene. Of these 1-hexene, 1-octene, 1-heptene, and 4-methyl-1-pentene are preferred. These  $\alpha$ -olefins can be used either individually or as a combination of two or more.

The amount of the unit derived from the  $\alpha$ -olefin preferably ranges from 5 to 30% by weight in the total copolymer.

In particular, the amount of said unit is more preferably from 10 to 25% by weight for obtaining a heat-activated label for use in the above-

described labeler adhesion method, and from 5 to 20% by weight for use in in-mold labeling.

If the amount of the unit derived from the α-olefin is lower than the above lower limit, the adhesive tends to exhibit insufficient adhesion, requiring high-temperature activation or molding. If it exceeds the above upper limit, the adhesive tends to cause blocking or blistering.

The catalyst system of the present invention can be a system consisting solely of a metallocene compound or a system comprising a metallocene compound as a main catalyst component along with a co-catalyst.

Useful metallocene compounds include those having at least one cyclopentadienyl skeleton as represented by formula (I):



wherein M represents a transition metal selected from the group consisting of Zr, Ti, Hf, V, Nb, Ta, and Cr; L is a ligand coordinating to the transition metal and represents a group having a cyclopentadienyl ring, a hydrocarbon group having 1 to 12 carbon atoms, an alkoxy group having 6 to 12 carbon atoms, an aryloxy group having 6 to 12 carbon atoms, a trialkylsilyl

group having 3 to 12 carbon atoms, an  $\text{SO}_3\text{R}$  group (wherein R represents a hydrocarbon group having 1 to 8 carbon atoms which may have a substituent, such as a halogen atom), a halogen atom or a hydrogen atom; x represents the same number as the valence of the transition metal; when a plurality of the ligands (L) coordinate, the L's may be the same or different provided that at least one of them is a cyclopentadienyl ring containing group: i.e., when x is 1, L is a group having a cyclopentadienyl ring; and when x is 2 or greater, at least one of the L's is a group having a cyclopentadienyl ring.

Illustrative examples of the group having a cyclopentadienyl ring are alkyl-substituted cyclopentadienyl groups, such as cyclopentadienyl, methylcyclopentadienyl, dimethylcyclopentadienyl, trimethylcyclopentadienyl, tetramethylcyclopentadienyl, pentamethylcyclopentadienyl, ethylcyclopentadienyl, methylethylcyclopentadienyl, propylcyclopentadienyl, methylpropylcyclopentadienyl, butylcyclopentadienyl, methylbutylcyclopentadienyl, and hexylcyclopentadienyl; an indenyl group, a 4,5,6,7-tetrahydroindenyl group, or a fluorenyl group. These groups may be substituted with a halogen atom, a trialkylsilyl group, etc.

Of the above-enumerated groups having a cyclo-

pentadienyl ring, an alkyl-substituted cyclopentadienyl group is particularly preferred.

Where the metallocene compound represented by formula (I) contains two or more groups having a cyclopentadienyl ring, two of the groups having a cyclopentadienyl ring may be linked together via an alkylene group, such as ethylene or propylene; a substituted alkylene group, such as isopropylidene or diphenylmethylenne; a silylene group; a substituted silylene group, such as a dimethylsilylene group, a diphenylsilylene group or a methylphenylenesilylene group.

The hydrocarbon group having 1 to 12 carbon atoms includes an alkyl group, such as methyl, ethyl, propyl, isopropyl or butyl; a cycloalkyl group, such as cyclopentyl or cyclohexyl; an aryl group, such as phenyl or tolyl; and an aralkyl group, such as benzyl or neophyl.

The alkoxy group includes groups such as methoxy, ethoxy, and butoxy. The aryloxy group includes groups such as phenoxy. The halogen atom includes fluorine, chlorine, bromine, and iodine.

The  $\text{SO}_3\text{R}$  group includes groups such as p-toluenesulfonato, methanesulfonato, and trifluoromethanesulfonato.

When M is a tetravalent transition metal, the

metallocene compounds containing a group having a cyclopentadienyl ring are represented by formula (II):



wherein M is a tetravalent transition metal;  $R^2$  represents a group (ligand) having a cyclopentadienyl ring;  $R^3$ ,  $R^4$ , and  $R^5$  each independently, represents a group having a cyclopentadienyl ring, a  $C_1-C_{12}$  alkyl group, a  $C_3-C_{12}$  cycloalkyl group, a  $C_6-C_{12}$  aryl group, a  $C_7-C_{12}$  aralkyl group, a  $C_1-C_{12}$  alkoxy group, a  $C_6-C_{12}$  aryloxy group, a  $C_3-C_{12}$  trialkylsilyl group, a  $SO_3R$  group (R being a substituted or unsubstituted hydrocarbon group having 1 to 8 carbon atoms), a halogen atom or a hydrogen atom; k represents an integer of 1 or greater; l, m and n each, independently, represent an integer of from 0 to 3, wherein  $k+l+m+n$  is equal to 4.

In the present invention, compounds represented by formula (II) in which at least two of  $R^2$ ,  $R^3$ ,  $R^4$ , and  $R^5$ , (e.g.,  $R^2$  and  $R^3$ ) are groups (ligand) having a cyclopentadienyl ring are preferably used. These groups having a cyclopentadienyl ring (e.g.,  $R^2$  and  $R^3$ ) may be linked together as described above.

Specific examples of the metallocene compound in which M is zirconium are shown below:

Bis(indenyl)zirconium dichloride, bis-(indenyl)zirconium dibromide, bis(indenyl)zirconium bis(p-toluenesulfonato), bis(4,5,6,7-tetrahydroindenyl)zirconium dichloride, bis(fluorenyl)zirconium dichloride, ethylenebis(indenyl)zirconium dichloride, ethylenebis(indenyl)zirconium dibromide, ethylenebis(indenyl)dimethylzirconium, ethylenebis(indenyl)diphenylzirconium, ethylenebis(indenyl)methylzirconium monochloride, ethylenebis(indenyl)zirconium bis(methanesulfonato), ethylenebis(indenyl)zirconium bis(p-toluenesulfonato), ethylenebis(indenyl)zirconium bis(trifluoromethanesulfonato), ethylenebis(4,5,6,7-tetrahydroindenyl)zirconium dichloride, isopropylidene(cyclopentadienylfluorenyl)zirconium dichloride, isopropylidene(cyclopentadienyl-methylcyclopentadienyl)-zirconium dichloride, dimethylsilylenebis(cyclopentadienyl)zirconium dichloride, dimethylsilylenebis(methylcyclopentadienyl)zirconium dichloride, dimethylsilylenebis(dimethylcyclopentadienyl)zirconium dichloride, dimethylsilylenebis(trimethylcyclopentadienyl)-zirconium dichloride, dimethylsilylenebis(indenyl)-zirconium dichloride, dimethylsilylenebis(indenyl)-zirconium bis(trifluoromethanesulfonato), dimethylsilylenebis(4,5,6,7-tetrahydroindenyl)zirconium dichloride, dimethylsilylenebis(cyclopentadienylfluor-

enyl)zirconium dichloride, diphenylsilylenebis-(indenyl)zirconium dichloride, methylphenylsilylene bis(indenyl)zirconium dichloride, bis(cyclopentadienyl)zirconium dichloride, bis(cyclopentadienyl)-zirconium dibromide, bis(cyclopentadienyl)methylzirconium monochloride, bis(cyclopentadienyl)ethylzirconium monochloride, bis(cyclopentadienyl)cyclohexylzirconium monochloride, bis(cyclopentadienyl)-phenylzirconium monochloride, bis(cyclopentadienyl)-benzylzirconium monochloride, bis(cyclopentadienyl)zirconium monochloride monohydride, bis(cyclopentadienyl)methylzirconium monohydride, bis(cyclopentadienyl)dimethylzirconium, bis(cyclopentadienyl)di-phenylzirconium, bis(cyclopentadienyl)dibenzylzirconium, bis(cyclopentadienyl)zirconium methoxychloride, bis(cyclopentadienyl)zirconium ethoxychloride, bis(cyclopentadienyl)zirconium bis(methanesulfonato), bis(cyclopentadienyl)zirconium bis(p-toluenesulfonato), bis(cyclopentadienyl)zirconium bis(trifluoromethanesulfonato), bis(methylcyclopentadienyl)zirconium dichloride, bis(dimethylcyclopentadienyl)zirconium dichloride, bis(dimethylcyclopentadienyl)zirconium ethoxychloride, bis(dimethylcyclopentadienyl)zirconium bis(trifluoromethanesulfonato), bis(ethylcyclopentadienyl)zirconium dichloride,

bis(methylethylcyclopentadienyl)zirconium dichloride,  
bis(propylcyclopentadienyl)zirconium dichloride,  
bis(methylpropylcyclopentadienyl)zirconium dichloride,  
bis(butylcyclopentadienyl)zirconium dichloride,  
bis(methylbutylcyclopentadienyl)zirconium dichloride,  
bis(methylbutylcyclopentadienyl)zirconium bis(methane-sulfonato), bis(trimethylcyclopentadienyl)zirconium dichloride, bis(tetramethylcyclopentadienyl)zirconium dichloride, bis(pentamethylcyclopentadienyl)zirconium dichloride, bis(hexylcyclopentadienyl)zirconium dichloride, and bis(trimethylsilylcyclopentadienyl)-zirconium dichloride.

In the above-enumerated zirconium compounds, the di-substituted cyclopentadienyl ring, such as dimethylcyclopentadienyl, can be a 1,2-substituted ring or a 1,3-substituted ring; the tri-substituted cyclopentadienyl ring, such as trimethylcyclopentadienyl, can be a 1,2,3-substituted ring or a 1,2,4-substituted ring; and the alkyl groups, such as propyl or butyl, can be branched or linear isomers, such as n-, i-, sec- and tert-alkyl groups. Compounds corresponding to the above-described zirconocene compounds, with the exception that the zirconium is replaced with titanium, hafnium, vanadium, niobium, tantalum or chromium, are also included in the metallocene compounds of the

present invention.

These metallocene compounds can be used either individually or as a mixture thereof. They may be used diluted with a hydrocarbon or a halogenated hydrocarbon.

Of the above-described metallocene compounds, zirconocene compounds having at least two groups having a cyclopentadienyl ring as ligands are preferred.

The co-catalyst which can be used in combination with the metallocene compound includes conventional aluminoxane compounds and compounds capable of reacting with the metallocene compound to form a stable anion.

The ethylene- $\alpha$ -olefin copolymer according to the present invention can easily be prepared in a conventional manner by using the above-described catalyst system. In detail, ethylene and an  $\alpha$ -olefin are copolymerized by liquid phase polymerization (such as solution polymerization or slurry polymerization), or gas phase polymerization in the presence of the catalyst system under a broad range of temperature and pressure conditions.

For example, polymerization in the presence of a combination of the metallocene compound and an aluminoxane compound can be carried out in accordance with

the processes described in Japanese Patent Laid-open Nos. 61-130314, 60-35006, 60-35007, 58-19309, and 60-35008 and Japanese Patent Laid-Open No. 3-163088, the relevant portions of which are hereby incorporated by reference. Polymerization in the presence of a combination of the metallocene compound and a compound capable of reacting with the metallocene compound to form a stable anion can be carried out in accordance with a high-pressure ionic polymerization process as described in European Patent No. 277,004 and Published International Patent Application No. WO 92/01723, the relevant portions of which are hereby incorporated by reference.

The melting point of MPE is preferably in the range of from 50° to 100°C, in terms of the temperature showing the peak of heat absorption for melting as measured with a differential scanning calorimeter (DSC) at a rate of temperature rise of 10°C/min.

In particular, the melting point of MPE for use in heat-activated labels to be used in a labeler adhesion method is preferably 50° to 80°C, and that for use in in-mold labeling is preferably 70° to 100°C.

The heat-activated label of the present invention is preferably one in which MPE has a melting point of 50 to 80°C and which is to be heated and then

stuck to a previously prepared molded article (heat-activated label for labeler adhesion) or one in which MPE has a melting point of 70 to 100°C and which is to be previously inserted into a mold for in-mold decoration (heat-activated label for in-mold decoration).

The number of melting points of MPE is arbitrary in agreement with the end use, i.e., one or more than one.

MPE preferably has a density of 0.86 to 0.93 g/cm<sup>3</sup>. In particular, the density is more preferably from 0.86 to 0.90 g/cm<sup>3</sup> in heat-activated labels for labeler adhesion, or from 0.88 to 0.92 g/cm<sup>3</sup> in heat-activated labels for in-mold decoration.

If the melting point or the density of MPE is lower than the respective preferred lower limits, the adhesive tends to cause blocking during storage or label release in the summer time. Further, the adhesive tends to have insufficient hot tack in heat-activated labels for labeler adhesion, or tends to cause blisters in labels for in-mold labeling. If these attributes exceed the respective preferred upper limits, the adhesive tends to exhibit insufficient adhesion.

MPE preferably has a melt index of 0.1 to 50 g/10 min., more preferably 0.3 to 30 g/10 min., as

measured in accordance with JIS K-7210 (190°C, 2.16 kg load). If the melt index is out of the above preferred range, the adhesive is not only difficult to process for example, in extrusion into an adhesive layer, but tends to exhibit insufficient adhesion.

In the present invention, MPE is preferably used in the form of a film. Therefore, MPE for use in the present invention is required to have satisfactory processability in various systems for monolayer or multilayer film formation by T-die extrusion, blown-film extrusion, and the like. With respect to this requirement, it is desirable for MPE to have either one of the following characteristics (1) and (2).

(1) To have longer branches than those formed by an  $\alpha$ -olefin. Existence of such long branches leads to improved melt properties and thereby to improved processability.

Ethylene- $\alpha$ -olefin copolymers having such long branches can be prepared by a conventional process as described in, PCT Application 093/08221 (corresponding to Japanese Patent Laid-Open No.7-500622 T2), the contents of which are hereby incorporated by reference. More specifically, ethylene is copolymerized with an olefin monomer and a vinyl compound other than the olefin monomer, said vinyl compound having a relatively

long hydrocarbon chain(C<sub>8</sub> or higher, preferably C<sub>16</sub> or higher) in the presence of a metallocene catalyst to obtain a desired branched polymer, i.e., an olefin polymer having long branches.

(2) To have two or more peaks in the molecular weight distribution or a relatively broad molecular weight distribution. Polymers having this characteristic contain low-molecular weight components and thereby exhibit improved processability.

Note that the low-molecular weight components referred to above are not those components that have such a low molecular weight that they cause blocking or blisters.

Ethylene- $\alpha$ -olefin copolymers having two or more molecular weight peaks or a relatively broad molecular weight distribution can be prepared by a conventional process as described in Japanese Patent Laid-Open No. 60-35008 or 7-500859 T2, the contents of which are hereby incorporated by reference. More specifically, they can be produced by (1) multi-stage polymerization, (2) polymerization using a mixed catalyst or (3) blending two or more resins having different molecular weight peaks.

A difference in melt viscosity with a dif-

ference in shear rate can be taken as an indication of the thus improved melt properties of the resin. The "difference" is a difference between the common logarithm of the viscosity in poise at a shear rate of  $10\text{ sec}^{-1}$ , which is referred to as  $\eta(10)$ , and the common logarithm of the viscosity in poise at a shear rate of  $100\text{ sec}^{-1}$ , which is referred to as  $\eta(100)$ , i.e.,  $\Delta \log \eta = \log \eta(10) - \log \eta(100)$ . The difference  $\Delta \log \eta$  of MPE is preferably 0.2 or more, more preferably 0.25 or more, most preferably 0.3 or more. With this condition being satisfied, further improved moldability can be obtained.

Compared with conventionally employed low-melting resins, MPE has a sharp molecular weight distribution and contains the comonomer component uniformly distributed in the polymer chain. By virtue of these attributes, MPE is characterized by the following: the melting point greatly drops linearly in direct proportion with an increase of the comonomer component; the content of low-molecular components that cause stickiness or blocking is extremely low; and high transparency is exhibited.

When used as an adhesive for labels, MPE offers the following advantages:

- 1) MPE displays sharp behavior in melting and

crystallization, which is advantageous for rapid heat activation and adhesion. In particular, since MPE has a small content of components that melt at temperatures below the melting point, blistering due to re-melting of the adhesive caused by heat-back (a phenomenon where the internal heat of a molded article removed from the mold is conducted to raise the surface temperature of the molded article) observed in in-mold labeling, can be inhibited. In the case of labeler adhesion, the sharp behavior in melting and crystallization is advantageous for rapid solidification.

2) The melting point can be controlled to conform with the molding temperature by adjusting the copolymerization ratio of ethylene and the  $\alpha$ -olefin. In other words, the heat activating temperature can be set arbitrarily according to the method of adhesion desired.

3) Because it comprises an  $\alpha$ -olefin as a comonomer, MPE exhibits high adhesion to olefin resin molded articles. Therefore, when the label is adhered to an olefin resin molded article, sufficient adhesion can be assured at a temperature lower than that required for a conventional adhesive having an equally low activating temperature.

4) If the proportion of a comonomer component in

conventional ethylene copolymers is increased for the purpose of reducing the melting point, the resulting copolymer has much increased levels of low-molecular components and therefore shows stickiness or causes blocking. To the contrary, MPE hardly causes blocking and has no stickiness due to the extremely small content of low-molecular components. Therefore, the heat-activated labels of the present invention have excellent processability in, various processes, such as printing, slitting or punching, and excellent handling properties in labeling operations, such as in inserting into a mold.

5) When a heat-activated label is required to be transparent, MPE can provide an adhesive layer free from haze. Thus, a label having extremely high transparency can be obtained.

MPE is commercially available under the trade names of AFFINITY (PL1845, PF114, SM1250, and HM1100), and ENGAGE (KC8852 and EG8200), all produced by Dow Chemical Co., Ltd.; EVOLVE (SP1540), produced by Mitsui Petrochemical Industries, Ltd.; and EXACT, produced by Exxon Corp. These products are suitable for use in the present invention.

The adhesive which can be used in the adhesive layer of the heat-activated label of the present invention

tion can be MPE alone or, if desired, a mixture of MPE and other substances.

Useful substances to be combined with MPE include polyethylene resins (such as low-density polyethylene, linear low-density polyethylene, high-density polyethylene, and copolymers of ethylene and 4-methyl-1-pentene, vinyl acetate, ethyl acrylate, acrylic acid, and methacrylic acid), polypropylene, polystyrene resins, polyester resins, polyamide resins, and petroleum resins.

When MPE is combined with three other substances, the other substances are preferably used in a proportion of not more than 50% by weight, more preferably not more than 30 wt% by weight, based on the total adhesive, so as not to impair the characteristics of MPE.

The adhesive layer can be made up solely of the above-described adhesive. If desired, the adhesive layer can also contain, in addition to the adhesive, various additives, such as plasticizers (such as liquid paraffin), waxes (such as paraffin wax and polyethylene wax), lubricants (such as fatty acid amides and fatty acid metal salts), anti-blocking agents (such as organic or inorganic fillers) so long as the expected effects of the invention are not impaired.

The adhesive layer preferably has a thickness of 0.5 to 50  $\mu\text{m}$ , more preferably 1 to 20  $\mu\text{m}$ .

While the heat-activated label of the present invention has a multilayer structure composed essentially of the above-described substrate and adhesive layer, it is usually printed to have a printed layer as stated previously. If desired, a conventional vacuum-evaporated metallic layer, a metal foil, a conventional adhesive layer for bonding two or more layers, or a conventional anchor coat layer, can also be provided. The substrate can be a single layer substrate or have two or more layers.

Layer structures generally used for the heat-activated labels of the present invention are, in order from the surface side: substrate/printed layer/vacuum-deposited metallic layer/adhesive layer, vacuum-deposited metallic layer/ printed layer/ substrate/ adhesive layer, first substrate/ printed layer/ vacuum-deposited metallic layer/second substrate/adhesive layer, and the like. If desired, an adhesive layer or an anchor coat layer can be provided between any two layers in these structures.

The heat-activated label of the present invention preferably has a total thickness of 30 to 300  $\mu\text{m}$ . If the thickness is smaller than 30  $\mu\text{m}$ , the label has

insufficient bending stiffness characteristics and is difficult to stick to a molded article or to insert into a mold. If the thickness exceeds 300  $\mu\text{m}$ , blisters tend to develop, particularly when the label is used in in-mold labeling.

The heat-activated label of the present invention can be easily produced as follows.

An adhesive layer and a substrate are laminated by co-extrusion, extrusion lamination (a method of extruding a molten adhesive on a substrate having been subjected to a preliminary surface treatment), dry lamination (a method of laminating a T-die extruded film of an adhesive or a blown film of an adhesive on a substrate via another adhesive), or other techniques, and other layers (such as a vacuum-deposited metallic layer or a printed layer) are then formed thereon by conventional methods. The other layers may be previously formed on the substrate prior to the laminating of the adhesive layer, if desired.

In the case of dry lamination, in order to obtain a label that does not suffer curling, it is preferable that a resin layer (second substrate), whose dynamic physical properties are similar to those of a substrate on which a film of adhesive is to be laminated (first substrate), is co-extruded together with

an adhesive to obtain a laminate film, which is then dry-laminated with the first substrate.

It is preferable to give an uneven pattern to the adhesive layer on the side to be brought into contact with a molded article (adherend) in order to avoid entrapping air between the adhesive layer and the molded article on labeling. Formation of an uneven pattern is particularly effective for heat-activated labels for in-mold labeling.

The uneven pattern may be regular (e.g., slits) or irregular. The size of the individual bosses or recesses may also be irregular. The height of the bosses is preferably 0.3 to 20  $\mu\text{m}$  and the area ratio of the bosses is preferably 30 to 90% based on the total area of the adhesive layer.

The uneven pattern can be formed, for example, by extruding a film of the adhesive on an engraved roll or embossing a molded film of the adhesive using an embossing roll. The uneven pattern can also be obtained by a method comprising forming a dot pattern made of a separate solvent-soluble hot melt adhesive (e.g., an ethylene-vinyl acetate copolymer) on the adhesive layer by gravure printing or a similar technique, or a method comprising adding particles of a filler, having a relatively large particle size to the adhesive layer.

The process for producing the labeled molded article according to the present invention will now be described in detail.

The first process according to the present invention is a process for producing a labeled molded article which comprises heating a heat-activated label of the present invention and sticking the heated label onto a previously prepared molded article (a labeler adhesion method).

Molding materials providing the molded article for use in the process of the present invention include polyolefin resins, such as ethylene resins and propylene resins, polystyrene resins, polyester resins, polyamide resins, and poly(meth)acrylate resins, with polyolefin resins being preferred. The molded articles to be labeled with the label of the invention include bottles, caps, sheets, films, tubes, pipes, and the like.

The first process preferably includes, a process comprising heating a heat-activated label by means of a heater to activate the adhesive layer and sticking the label while in an activated state onto a molded article; and a process, as illustrated in Fig. 1, comprising fixing a heat-activated label 1 on a heated support 12 with the adhesive layer facing out-

side, bringing the label, as fixed on the support 12 and activated by the heat of the support 12, into contact with a bottle 11 (a molded article), and transferring and adhering the label 1 to the bottle 11. The support 12 is preferably a roll.

For securing high adhesive strength and reducing the time for adhesion, it is preferable to set the heat activating temperature as high as possible within a range that does not cause deformation of the molded article.

The second process of the present invention is a process for producing a labeled molded article comprising producing a molded article by using a mold into which a heat-activated label of the present invention has previously been inserted(in-mold labeling).

The molded article according to the second process is preferably a blown container (i.e., a hollow container obtained by blow molding). In the blown container, at least the side to which the label is to be stuck comprises a polyolefin resin.

The blown container includes a multilayer blown bottle having, for example, an ethylene resin layer as an outermost layer and a barrier layer made of an ethylene-vinyl alcohol copolymer resin as an inner layer.

The mold to be used in in-mold labeling can be any mold that is conventionally used in the production of molded articles.

In-mold labeling can be carried out in any manner so long as molding is conducted using a mold previously containing the label inside. For example, in-mold labeling includes, but is not limited to, blow molding (inclusive of extrusion blow molding, injection blow molding, etc.), compression molding, pressure forming, vacuum forming, and injection molding.

The heat-activated label can be fixed to a cavity wall in a conventional manner, for example, by suction or static electricity.

The resin temperature in the above-described in-mold labeling depends on the resin being molded. For example, the temperature of polyethylene is 160 to 240°C in extrusion blow molding, 180 to 260°C in injection molding, and 160 to 240°C in pressure forming and vacuum forming. In order to minimize the molding cycle, the resin temperature is preferably set low within the above ranges. The molding pressure is 1.5 to 15 kgf/cm<sup>2</sup> in extrusion blow molding and pressure forming; 20 to 200 kgf/cm<sup>2</sup> in injection molding; and 2 to 600 mmHg in vacuum forming. Taking cooling efficiency into account, the pressure of pressing the resin

onto the mold is preferably set high within the above ranges.

Extrusion blow molding as an embodiment of the second process is set forth below in detail by referring to the drawings. As shown in Fig. 2(a), resin is extruded through a die 21 to form a parison 22. A heat-activated label 1 has been fixed in a mold 23. As shown in Fig. 2(b), the mold 23 is closed, and air is blown into the inside of the parison 22 at the above-required pressure to blow the parison 22 into a container 24 and, at the same time, to adhere the label 1 to the outer surface of the container 24.

#### EXAMPLES

The present invention will now be illustrated in greater detail by way of Examples and Comparative Examples, but it should be understood that the invention is not deemed to be limited thereto.

Resins used as an adhesive in the Examples and Comparative Examples are shown in Table 1 below. Table 1 also shows  $\Delta \log \eta$ , an indication of molding properties, of the ethylene-a-olefin copolymers used in the Examples.

TABLE 1

Abbreviation	Resin	Maker	Trade Name	M I (g/10min)	Density (g/cm <sup>3</sup> )	Melting Point (°C)	Δ log η
MPE 1	Metallocene LDPE **	Dow Chemical Co. Ltd.	PL1845	3.5	0.910	103	0.33
MPE 2	Metallocene LDPE **	Mitsui Petro chemical Industries, Ltd.	SP1540	1	0.915	97, 112	0.25
MPE 3	Metallocene LDPE **	Dow Chemical Co. Ltd.	PF114	1.6	0.895	93, 80	0.47
MPE 4	Metallocene LDPE **	Dow Chemical Co. Ltd.	SM1250	3.0	0.885	78, 65	0.42
MPE 5	Metallocene LDPE **	Dow Chemical Co. Ltd.	HM1100	1.8	0.880	71, 58	0.40
MPE 6	Metallocene LDPE **	Dow Chemical Co. Ltd.	KC8852	3	0.875	66, 51	0.37
MPE 7	Metallocene LDPE **	Dow Chemical Co. Ltd.	EG8200	5	0.870	61, 49	0.46
EVA 1	Ethylene-vinyl acetate copolymer	Mitsubishi Chemical Corporation	LV250	3	0.93	103	—
EVA 2	Ethylene-vinyl acetate copolymer	Mitsubishi Chemical Corporation	LV350	3.5	0.93	92	—
EVA 3	Ethylene-vinyl acetate copolymer	Mitsubishi Chemical Corporation	LV570	1.5	0.94	81, 65	—
EVA 4	Ethylene-vinyl acetate copolymer	Mitsubishi Chemical Corporation	LV670	1.5	0.95	67	—
EMAA	Ethylene-methacrylic acid copolymer	Du Pont-Mitsui Poly chemical Co., Ltd.	108C	8	0.94	96	—

Note: \* In a descending order (for a plurality of peaks).

\*\* Ethylene-*a*-olefin copolymer prepared by copolymerization using a metallocene compound-containing catalyst system.

EXAMPLE 1

Labeler Adhesion Method

A heat-activated label was prepared as follows.

Homopolypropylene (PP) (B200, a trade name, produced by Mitsui Petrochemical Industries, Ltd.) and the adhesive shown in Table 2 below were co-extruded through a T-die into a double-layer laminate film to obtain a laminate composed of a substrate and an adhesive layer. For cooling of the extruded film, a cooling roll having an uneven surface was used for the adhesive layer side to give to the adhesive layer a fine uneven pattern comprising bosses having a height of 3  $\mu\text{m}$  and an area ratio of 70% based on the total area.

The substrate was 100  $\mu\text{m}$  thick, and the adhesive layer was 10  $\mu\text{m}$  thick.

After printing the substrate of the laminate film, the film was punched into 120 mm long and 60 mm wide heat-activated labels according to the present

invention.

As an adherend, a high-density polyethylene (HDPE) blown container (about 200 mm high, having an approximately elliptical section of about 90 mm in major axis and 75 mm in minor axis) produced in a conventional extrusion blow molding machine was prepared.

The heat-activated label was fixed to a roll set at 80°C with the adhesive layer outside, and the label, activated by the heat of the roll, was transferred and adhered to the container.

The same labeling operation was conducted at a roll temperature of 100°C. "100°C" was the upper limit of the roll temperature because the container sometimes undergoes deformation at label temperatures higher than 110°C.

The adhesive strength of the label on the container was measured according to the following test method. The result obtained is shown in Table 2.

Measurement of Adhesive Strength:

The container with the label adhered thereto was cut into a 15 mm wide strip and subjected to a tensile test at a rate of pulling of 300 mm/min at 23°C. The peel strength of the label from the container was taken as an adhesive strength.

Practical adhesive strength varies depending on the use. An adhesive strength of at least 300 gf/15 mm is required for those labels which do not need to be removed afterward, and at least 100 gf/15 mm for stripable labels.

EXAMPLES 2 TO 4 AND COMPARATIVE EXAMPLES 1 TO 4

Heat-activated labels and labeled molded articles were prepared in the same manner as in Example 1, except for using the resins shown in Table 2 below as an adhesive. The adhesive strength of the resulting labels was measured in the same manner as in Example 1. The results obtained are shown in Table 2.

TABLE 2

		Adhesive	Adhesive Strength (gf/15mm)	
			Roll Temperature: 80°C	Roll Temperature: 100°C
Example No.	1	MPE 4	90	370
	2	MPE 5	130	400
	3	MPE 6	100	520
	4	MPE 7	170	490
Comparative Example No.	1	EVA 2	10	150
	2	EVA 3	30	200
	3	EVA 4	20	170
	4	EMMA	0	20

As is apparent from the results in Table 2, the heat-activated labels according to the present invention firmly adhere to the molded article under either labeling condition, exhibiting surprisingly higher adhesiveness than the comparative labels. Every comparative label exhibited insufficient adhesion at an adhesion temperature of 80°C, causing appreciable blisters at the edge area and having very low adhesive strength. In particular the label using EMMA (a copolymer of ethylene and methacrylic acid) having the highest polarity of the adhesives used in the Examples

and Comparative Examples developed considerable blisters at either adhesion temperature, exhibiting a state far from adhesion.

#### EXAMPLE 5

##### In-Mold Labeling

A heat-activated label was prepared as follows.

A 50  $\mu\text{m}$  thick PP film (SI-60, a trade name, produced by Tohcello Co., LTD.) was printed by gravure printing to obtain a laminate film having a printed layer (hereinafter referred to as laminate A).

Separately, PP (B200, a trade name, produced by Mitsui Petrochemical Industries, Ltd.) and the adhesive shown in Table 3 below were co-extruded through a T-die to obtain a double-layered laminate film composed of the 50  $\mu\text{m}$  thick PP film as a substrate and an adhesive layer having a thickness of 10  $\mu\text{m}$  (hereinafter referred to as laminate B). For cooling of the extruded film, a cooling roll having an uneven surface was used for the adhesive layer side to give to the adhesive layer a fine uneven pattern comprising bosses having a height of 3  $\mu\text{m}$  and an area ratio of 70% based on the total area.

The laminates A and B were adhered into a single laminate film, with the adhesive layer of the

former and the substrate of the latter facing each other, by dry lamination via a urethane type adhesive. The resulting laminate film was cut into 120 mm long and 60 mm wide labels of the present invention.

Hollow containers labeled with the above prepared heat-activated label were produced by extrusion blow molding as follows.

High-density polyethylene (5503D, a trade name, produced by Showa Denko, K.K.) was fed to an extrusion blow molding machine and extruded to form a tubular parison of a molten resin (180°C). Compressed air (5 kgf/cm<sup>2</sup>) was blown into the hot parison in a mold in which the label had been inserted to obtain a labeled blown container.

The cooling time in the mold was 10 seconds.

Labeled blown containers were produced in the same manner as described above, except for changing the cooling time to 8 seconds or 6 seconds.

The resulting labeled blown containers were evaluated in terms of adhesive strength and blistering as follows. The results of the evaluation are shown in Table 3.

Adhesive Strength:

Evaluated in the same manner as in Example 1.

Blister:

The label was observed with the naked eye at about 24 hours after molding and graded according to the following standard.

A ... No blister was observed.

B ... There were blisters which were nearly imperceptible at a glance.

C ... There were blisters perceptible at a glance.

EXAMPLES 6 TO 9 AND COMPARATIVE EXAMPLES 5 TO 8

Heat-activated labels and labeled molded articles were prepared in the same manner as in Example 5 except for using the resins shown in Table 3 as an adhesive. The resulting labeled containers were evaluated in the same manner as in Example 5. The results obtained are shown in Table 3.

TABLE 3

	Adhesive	Adhesive Strength (gf/15mm)	Blisters		
			Cooling Time		
			10 sec	8 sec	6 sec
Example No.	5 MPE 1	300	A	A	C
	6 MPE 2	340	A	A	C
	7 MPE 3	4000 or more	A	A	A
	8 MPE 4	4000 or more	A	A	A
	9 MPE 5	4000 or more	A	A	A
Comparative Example No.	5 EVA 1	60	C	C	C
	6 EVA 2	280	C	C	C
	7 EVA 3	1200	A	C	C
	8 EMAA	20	C	C	C

Note: \* Measured on samples obtained by 6-second cooling.

Examples 5 and 6 offer instances of using, as an adhesive, metallocene LLDPE whose melting point is relatively high, i.e., about 100°C. The labels of Examples 5 and 6 had higher adhesive strengths than the labels of Comparative Examples 5 and 6 which used EVA whose melting point is lower than that of metallocene

LLDPE, exhibiting sufficient strength for practical use. Although blisters occurred in the case of 6-second cooling, no blisters was observed in the case of 8-second cooling, proving that these labels are practically useful.

Examples 7 and 8 afford instances in which metallocene LLDPE having a lower melting point than that used in Examples 5 or 6 was used as an adhesive. In the adhesive strength test, interlaminar peeling between the adhesive layer and the substrate or a failure of the substrate occurred, and the measured adhesive strength between the adhesive and the container was as high as more than 4000 gf/15 mm. No blister was produced for either cooling cycle.

Comparative Examples 5 to 7 show the results of using EVA having different melting points as an adhesive. Compared with metallocene LLDPE having equal melting points, these adhesives had lower adhesive strengths and caused blisters, except in the case when cooling was sufficiently conducted in Comparative Example 7.

In Comparative Example 8 using EMMA having the highest polarity of all the other adhesives used in the Examples and Comparative Examples, the label not only had an extremely small adhesive strength but developed

large blisters at any cooling time.

This application is based on Japanese Patent Application No.8-35210, filed with the Japanese Patent Office on February 22, 1996, the entire contents of which are hereby incorporated by reference.

Obviously, additional modifications and variations of the present invention are possible in light of the above teachings. It is therefore to be understood that within the scope of the appended claims, the present invention may be practiced otherwise than as specifically described herein.

CLAIMS:

1. A heat-activated label comprising a substrate having a front and a back and an adhesive layer provided on the back of the substrate, wherein said adhesive layer comprises an ethylene- $\alpha$ -olefin copolymer obtained by copolymerization of ethylene and an  $\alpha$ -olefin in the presence of a catalyst comprising a metallocene compound.
2. The heat-activated label as claimed in claim 1, wherein said ethylene- $\alpha$ -olefin copolymer has a melting point of 50 to 80°C.
3. The heat-activated label as claimed in claim 2, wherein said ethylene- $\alpha$ -olefin copolymer has a melting point of 70 to 100°C.
4. The heat-activated label as claimed in any preceding claim, wherein the substrate has a thickness of 40 to 200  $\mu$ m.
5. The heat-activated label as claimed in any preceding claim, wherein the  $\alpha$ -olefin is an  $\alpha$ -olefin having from 3 to 20 carbon atoms.
6. The heat-activated label as claimed in claim 5,

wherein the  $\alpha$ -olefin is at least one member selected from propylene, 1-butene, 1-pentene, 1-hexene, 1-octene, 1-heptene, 4-methyl-1-pentene, 4-methyl-1-hexene, 4,4-dimethyl-1-pentene, octadecene and iso-octene.

7. The heat-activated label as claimed in any preceding claim, wherein the ethylene- $\alpha$ -olefin copolymer comprises from 5 to 30% by weight of the unit derived from the  $\alpha$ -olefin.

8. The heat-activated label as claimed in claim 7, wherein the unit derived from the  $\alpha$ -olefin is present in the ethylene- $\alpha$ -olefin copolymer in an amount of from 10 to 25% by weight.

9. The heat-activated label as claimed in claim 8, wherein the unit derived from the  $\alpha$ -olefin is present in the ethylene- $\alpha$ -olefin copolymer in an amount of from 5 to 20% by weight.

10. The heat-activated label as claimed in any preceding claim, wherein the metallocene compound is a compound of formula (I).

$ML_x$  (I)

wherein M is a transition metal selected from Zr, Hf, Ti, V, Nb, Ta, and Cr; L is a ligand selected from groups having a cyclopentadienyl ring, C<sub>1</sub>-C<sub>12</sub> hydrocarbon groups, C<sub>1</sub>-C<sub>12</sub> alkoxy groups, C<sub>6</sub>-C<sub>12</sub> aryloxy groups, trialkylsilyl groups having 3 to 12 carbons, -SO<sub>3</sub>R groups, halogens and hydrogen; wherein R is a C<sub>1</sub>-C<sub>8</sub> hydrocarbon group that is unsubstituted or substituted; and x is the valence number of the metal M; wherein at least one group L must be a group having a cyclopentadienyl ring.

11. The heat-activated label as claimed in any of claims 1 to 9, wherein the metallocene compound is a compound of formula (II).



wherein M is a tetravalent transition metal; R<sup>2</sup> is a group having a cyclopentadienyl ring; each of R<sup>3</sup>, R<sup>4</sup> and R<sup>5</sup> is, independently, a member selected from hydrogen, halogens, cyclopentadienyl ring containing groups, C<sub>1</sub>-C<sub>12</sub> alkyl groups, C<sub>3</sub>-C<sub>12</sub> cycloalkyl groups, C<sub>6</sub>-C<sub>12</sub> aryl groups, C<sub>7</sub>-C<sub>12</sub> aralkyl groups, C<sub>1</sub>-C<sub>12</sub> alkoxy groups, C<sub>6</sub>-C<sub>12</sub> aryloxy groups, C<sub>3</sub>-C<sub>12</sub> trialkylsilyl groups and -SO<sub>3</sub>R groups, wherein R

is a substituted or unsubstituted hydrocarbon group having from 1 to 8 carbons; k is an integer of 1 or greater; l, m and n are each, independently, an integer from 0 to 3, wherein  $k+l+m+n=4$ .

12. The heat-activated label as claimed in any preceding claim, wherein the metallocene compound is a zirconoene compound.

13. The heat-activated label as claimed in any preceding claim, wherein the ethylene-a-olefin copolymer has a density of from 0.86 to 0.93 g/cm<sup>3</sup>

14. The heat-activated label as claimed in claim 13, wherein the ethylene-a-olefin copolymer has a density of from 0.86 to 0.90 g/cm<sup>3</sup>.

15. The heat-activated label as claimed in claim 13, wherein the ethylene-a-olefin copolymer has a density of from 0.88 to 0.92 g/cm<sup>3</sup>.

16. A process for producing a labeled molded article, which comprises heating a heat-activated label as claimed in claim 1, and applying the heated heat-activated label to a surface of a molded article.

17. The process as claimed in claim 16, wherein said ethylene-a-olefin copolymer has a melting point of 50 to 80C°.

18. A process for producing a labeled molded article, which comprises producing a molded article in a mold, wherein a heat-activated label as claimed in any of claims 1 to 15 is inserted into the mold prior to producing the molded article.

19. The process as claimed in claim 18, wherein said ethylene-a-olefin copolymer has a melting point of 70 to 100C°.

20. The process for producing a labeled molded article according to claim 19, wherein said molded article is a blown container, wherein at least a side to which the label is to be applied comprises a polyolefin resin.

21. Use of the heat-activated label of claim 1 as a label for production of labeled molded articles.



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**Claims searched:** 1 to 21

**Examiner:** R.J.MIRAMS  
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**Patents Act 1977**  
**Search Report under Section 17**

**Databases searched:**

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.O): B5N

Int Cl (Ed.6): B32B 7/12, 27/04, 27/32. C09J 123/00, 123/02, 123/04, 123/08, 123/16.  
G09F 3/04.

Other: ONLINE: WPI, CLAIMS.

**Documents considered to be relevant:**

Category	Identity of document and relevant passage	Relevant to claims
A	WO 92/12212A1 (Exxon)	
A	US 5,530,054A (Tse)	

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
& Member of the same patent family		E	Patent document published on or after, but with priority date earlier than, the filing date of this application.